Author responses to reviewers' comments of acp-2020-1171 (*COVID-19 lockdowns highlight a risk of increasing ozone pollution in European urban areas*)

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Response to reviewers

Anonymous Referee #1

This paper analyzed the effect of the European COVID-19 lockdowns on NO_2, O_3, and O_x concentrations by comparing the observation and business as usual (BAU) derived from machine learning at 246 stations. The lockdown effect was determined by the Bayesian change point models. This analyze gave an 34% reduction of NO_2 concentration and 30% increase of O_3 leading to little change in O_x. Therefore, the change in NO_2 and O_3 is mainly a repartitioning of O_x. This paper presents a timely and important analysis of evaluating the lockdown impact on air quality in Europe. The paper is well written and structured. I suggest the authors to consider the following comments, which may help to improve the paper.

Thank you for your positive comments and suggestions for improvements. Please see the itemised responses below.

General comments

1. The description of methods are not detailed enough. Although most of the methods are used and described in previous studies, more detail information, for example about how the calculation of BAU, is useful for the reader to understand the data processing. There are some references provided to show how to perform the data analysis but are written in a specific programming language. The fundamental description in the paper would be helpful in case some of the readers are not using this tool.

   We have expanded the methods section to describe the method's approach further. The previous papers we have references give comprehensive details on the methods used and therefore, here, we focus on explaining the calculation of the business as usual scenario. The new text now reads:

   "The philosophy of this approach involves using a machine learning model, trained on past data, to predict beyond the last observations it has seen. The model is trained on a long enough period, two years in this work, to capture the variability of concentrations experienced in a variety of meteorological conditions. Beyond the training period (February, 14, 2020), the model predicts concentrations based on meteorological variables which from the model's perspective are from the future. The time series which results is a *counterfactual*. This counterfactual represents an estimate of concentrations during a business as usual (BAU) scenario. The BAU concentrations can be readily compared with what was observed for example, Figure 3 and the changes quantified, explained, and interpreted. This allows for a robust comparison with what was expected with what was observed."

2. The input and output of random forest model is hourly data. This is different from Grange et al. (2018), where daily averaged data was used. Why is the change? If hourly data is used, I am not sure how well the model captures the lockdown effect if the variability is mainly contributed by diurnal variation. Also, it would be very helpful to show the performance of the BAU calculation in a time series plot in their absolute concentration, perhaps in the supplement. The current comparison is only show in Fig A1 with some averaged R^2 is not enough. At least the performance for different countries should be show individually unless the model performances are the same. The calculation of O_x BAU is not clear. Is it calculated from the O_x observation like NO_2 and O_3, or the sum of NO_2_BAU and O_3_BAU?

   Hourly data were used because (a), they result in more performant models when compared to lower resolution models, chiefly because explanatory variables such as wind direction have far more information at higher resolution and (b), these hourly data are available for the analysis time period. Grange et al. (2018) analysed PM_{10} data over a much longer period and these data were collected by gravimetric samplers and therefore, were at daily resolution.

   We agree that additional O_3 metrics (such as rolling 8-hour means) might capture some attributes which maybe somewhat hidden by the mean response. However, we would argue that the mean response of all species is the most important metric and will represent the changes in concentrations well.

   Displaying the sites' time series is not practical in a publication such as this. However, we have provided an example time series in concentration units in Figure 3. Figure 3 very clearly demonstrates the counterfactual divergence from the observed concentrations.

   We agree that the lack of model error statistics was a weakness in the manuscript. We have addressed this by replacing Figure A1 with a more comprehensive version (also below in Figure 1). The new figure shows Pearson's correlation coefficient (r), mean bias

(MB), normalised mean bias (NMB), and normalised root mean square error (NRMSE) for all sites' models for the training and validation periods.

The O_x modelling was indeed done with observational data like NO_2 and O_3. This statement was missing in the manuscript and has been edited:

"... random forest models were trained to explain hourly mean NO_2, O_3, and O_x concentrations using surface meteorological and time explanatory variables for each monitoring site."

3. The argument of ozone pollution need for evidence to support. As discussed in section 3.5, the increase in O_3 is mainly a repartitioning of O_x during the lockdown. The $O_x/O_3/NO_2$ concentrations were missing so I cannot tell from the paper itself if all O_x are in the form of O_3, will O_3 exceed the limit? This is a rough estimation assuming only repartitioning play a role. As mentioned in the paper, the ozone formation is nonlinear with VOC and NO_x and Europe is likely in the VOC-limited regime. Reduction in NO_x do not lead to higher O_3 formation. If the reduction in NO_x is stronger than lockdown in the future, ozone production could move to NO_x-limited regime, which ozone pollution less important.

   The reviewer rightly emphasizes the non-linear nature of its production involving NO_x and VOCs. However, the focus of the current study is constrained to urban areas including roadside locations, rather than regional scale rural locations. To understand this issue more fully at a European scale would require air quality modelling. However, as shown in Figure 6, O_x concentrations varied little at background and traffic sites in comparison with either NO_2 or O_3. As discussed elsewhere in the paper, (and shown Figure 6), there is more evidence of O_x concentrations decreasing at traffic sites, which can be attributed to reductions in the primary emission of NO_2.

Technical comments

1. Line 109: The model prediction is corrected by -3.7ug for NO_2. How much you result sensitive to this correction.

   We apologise, but we do not understand this question. However, we believe that what is being asked is "how sensitive are your results to the bias correction applied to NO_2".

   The correction applied resolved the systematic underprediction of NO_2 due to already lower emissions before the lockdowns were officially implemented — see response to Line 112 below where the use of a correction is used primarily to support a consistent representation of changes relative to a specific date.

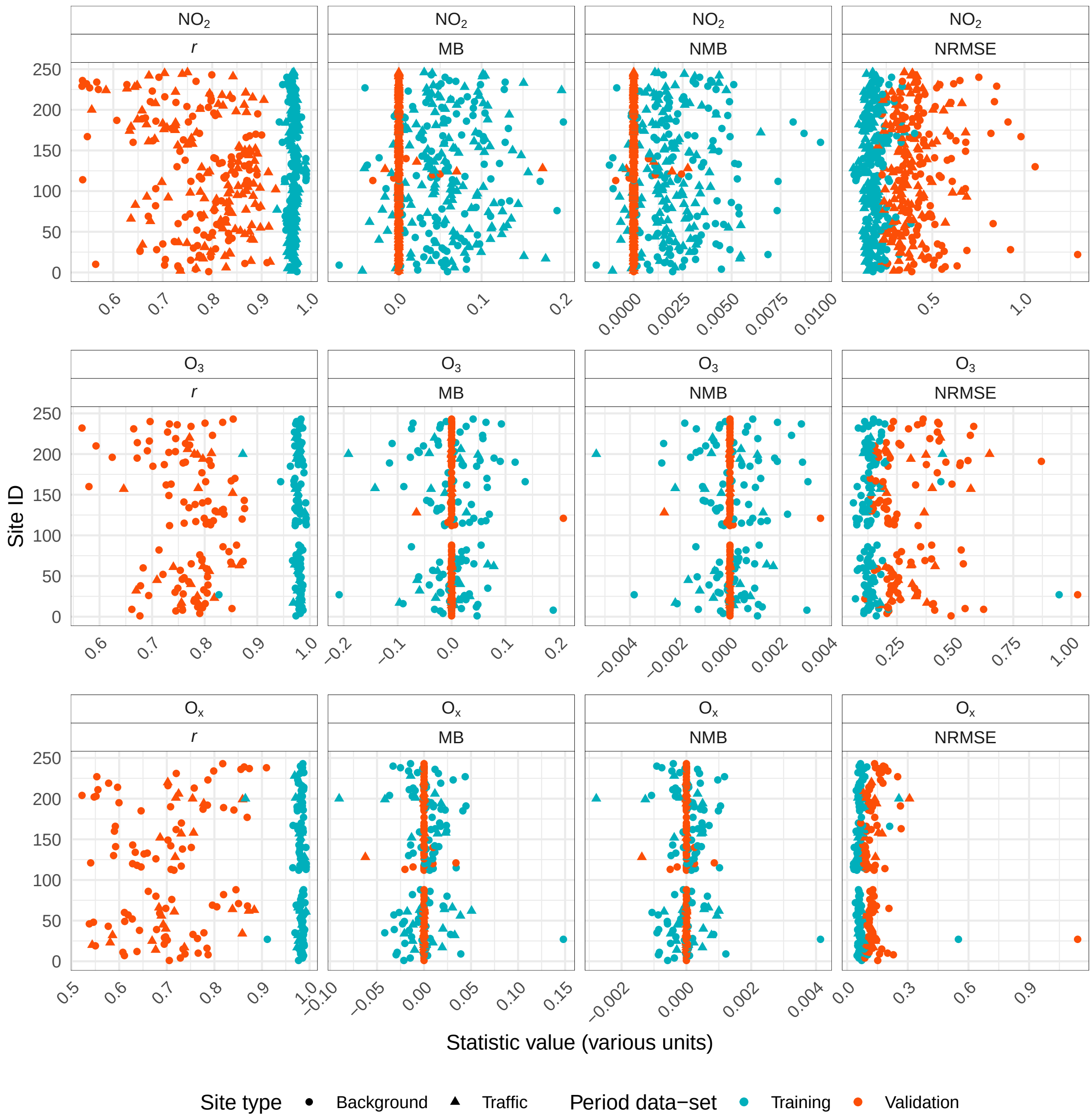

Figure 1: Model error summaries for all monitoring sites' (coded as integers) NO_2, O_3, and O_x models for two datasets – the training and validation sets. The error summaries are Pearson's correlation coefficient (r), mean bias (MB; in $\mu g\,m^{-3}$), normalised mean bias (NMB), and normalised root mean square error (NRMSE). The normalised were normalised by the observed mean.

2. Line 112: The underprediction of NO_2 is attributed to mild temperature and windy conditions. Isn't this indicating the model is not able to predict the condition in 2020?

   A substantial component of the under prediction would be due to already curtailed economic activity and therefore, emissions before the introduction of lockdowns, and is primarily used to provide a consistent basis for quantifying the changes relative to a specific point in time. The bias correction mostly resolved these issues and the model performance was very good (see Figure A1). The random forest modelling approach used has limitations on what it can achieve. The under prediction of NO_2 2020 can be partially explained by the rather unusual weather conditions experienced in the spring of 2020 which the model could not represent perfectly.

3. Line 210–213: The projection of NO_2 reduction and O_3 increase in the future is assuming a linear trend, which seems to me a bit too simple. Especially calculating the year to reach the lockdown impact bothers me.

   The goal of these calculations and discussion was put the changes observed in 2020 into context – they do not have the objective of being predictions. We have added a sentence explicitly stating this:
   "These calculations have not been done to predict future concentrations, only to put the changes experienced between March and July, 2020 in context."
   The abstract has also been slightly edited for clarity around this point.

4. Line 229–230: This sentence is not clear.

   We have edited for this sentence for clarity and it now reads:
   "The roadside increment in NO_2 above urban background concentrations diminished considerably over lockdown due to large reductions in vehicle activity."

5. Line 233: maybe you want to refer your argument to table 1.

   Done

6. Line 274: I think better to state the access of both NO and NO_2 data.

   Done.

Referee: Shaojun Zhang Referee #2

Grange et al. utilized time-series random forest models to analyze the changes of NO_2 and O_3 concentrations caused by the COVID-19 lockdowns across European countries. This work has important findings from the natural experiment of atmospheric pollution that most urban areas in Europe is in the VOC-limited scheme of O_3 formation (e.g., at least in Spring). Therefore, only mitigating traffic NO_x emissions might bring in unwanted increase of urban O_3. Overall, the manuscript is well organized, and the data analysis is solid and consistent.

Thank you for your positive comments. Please see the itemised responses below.

1. Line 29: I suggest add the explanation of the evaluation metric of Google mobility; e.g., the search frequency of points of interest, or the visit frequency (or duration spent) at points of interest?

   The Google mobility data is highly anonymised and only reports "movement trends" in contrast to a baseline. The text and the figure caption has been altered to clearly explain this:

   "Google's mobility data (Google, 2020) based on movement trends very effectively demonstrates the change in mobility based on a baseline (Figure 1)."

2. Line 36: Please reconsider the wording "near-minimum". I suppose commercial, transportation and recreation activities would be drastically declined, and the impact on essential industrial sectors would be less substantial.

   The text has been altered and now reads:

   "The European lockdowns can be thought of and approached as an air quality 'experiment' where economic activity was substantially curtailed where commercial, transportation, and recreation activities drastically declined."

3. Line 57: Please describe the distance between traffic sites and urban-BG sites in the selected urban areas. I wonder whether these traffic sites in various European countries would be deployed based on a unified, clear principle (e.g., distance to road curb, daily traffic volume)? Or, consider to enhance the statement around Line 70.

   We have calculated the distances among the different urban-traffic and urban-background sites within each urban area. The mean and median distances among the sites in an urban area was 5.2 and 3.9 km respectively. Therefore, the majority of the sites were in rather close proximity to one another and offer good comparisons to one another. The text has been updated to reflect this:

"The mean distance among the different air quality monitoring sites within an urban area was 5.2 km."

The site-type classification was done on the data contained in the European Air Quality e-Reporting (AQER) database. Although the classifications of the sites may contain differences among the authorities supplying data to AQER database, efforts are made to apply the same classification system among all member states.

4. Line 65: Please briefly describe how to match air quality and weather sites in this study.

   The text has been updated to describe the matching logic:

   " The matching logic between the air quality and meteorological sites was simple. The nearest ISD site to a particular air quality site was determined, the observations queried, and tested to ensure the data record was complete for the analysis period."

5. Line 104: It is not clear, in Figure A1, whether the distribution of R^2 represents the interval of R^2 (minimum to maximum) for each site-specific RF model? In addition to R^2, other validation metrics like normalized mean error can be used to evaluate the average discrepancy between modelled and observed results. And, I am surprised that both NO_2 and O_3 share good model validation results but O_x has lower R^2. What are the possible reasons and implications?

   Figure A1 has been replaced with a more comprehensive version to address the limited information supplied in the original manuscript (see Figure 1 and Referee #1's second general comment). The updated version displays more metrics, including normalised mean bias and normalised root mean square error.

   It is indeed interesting to see that the predictive performance of O_x is somewhat lower than NO_2 and O_3. This can be explained by O_x displaying less of a diurnal cycle than other pollutants. The somewhat invariant concentrations result in the "hour" variable having less information gain when compared to the other modelled variables.

6. Line 109: what is the percentage of underestimation.

   The mean percentage change has now been added to the text:

   "The under-prediction was on average, -3.7 $\mu g\,m^{-3}$ (95 % CI: [-4.2, -3.3]; mean percentage change: 15.9 %)."

7. Line 147: What is the possible cause (from the perspective of atmospheric chemistry or model validation performance) of comparable O_3 concentrations in the late period of this analysis to the business-as-usual levels, while NO_2 concentrations still indicated some degree of NO_x emission reduction?

We believe that actual NO_2 and NO_x emissions remained lower than the business-as-usual scenarios across Europe until the end of July, 2020. The difference between the observed and predicted NO_2 concentrations at traffic sites was greater than the urban-background sites. This suggests that traffic emissions are mostly responsible for this. Despite the reduction in emissions, there was still adequate NO_x in European urban atmospheres to generate business-as-usual O_3 concentrations by the end of the analysis period (July 31, 2020).

8. Line 170: I consider the less correlated relationship between lockdown date and O_3 surge possible is because O_3 is a more regional pollutant than NO_2 (high contribution from regional transport). I wonder how about analyzing the maximum daily average 8-hr instead of all O_3 observations?

   We also believe that analysing additional O_3 metrics may provide some value. However, analysing the mean response is the most useful and provides consistency with the other species included in the analysis. The extension of the analysis to additional O_3 metrics would be better handled in future work and studies.

9. Line 185: Is there any supporting mobility data to verify the actual change of mobility activities in Germany and Switzerland vs. in France and Italy?

   The stringency indices (Figure A2) indicate that the countries such as France and Italy had a more strict collection of policies when compared to other countries such as Germany and Switzerland. The © Google's mobility also support this (Figure 2).

10. Line 205: Please consider to add the increase of maximum daily average 8-hr ozone concentrations.

    Please see our above response to question/point 8.

11. Line 210: The authors has strong assumptions that the future reduction pace of NO_2 would follow that in the past decade, and the O_3 increase would greatly relate to the change of traffic emissions. I am not very confident with these assumptions. In particular, O_3 pollution is a regional issue, and is relevant to emission controls not only for NO_x but also for VOCs (e.g., deeper mitigation of NO_x might lead to O_3 reduction). Similar concern for the statement in the abstract (e.g., the predicted situation in 2028).

    The same question has been asked by Referee #1 (technical comment 3). The goal of these calculations and discussion was put the changes observed in 2020 into context – they do not have the objective of being predictions. We have added a sentence explicitly stating

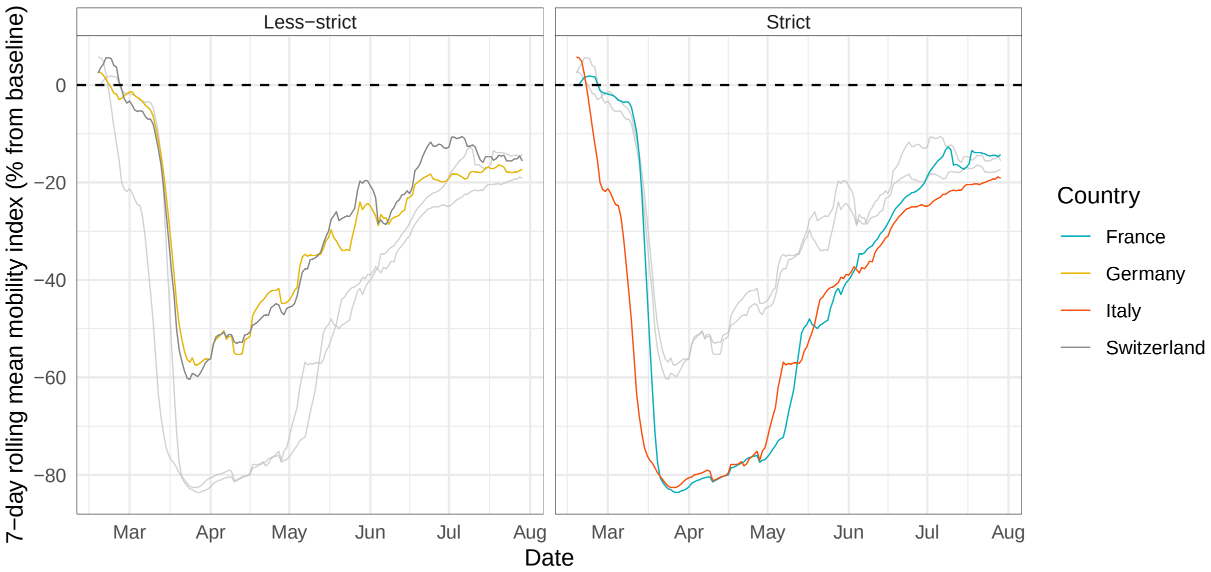

Figure 2: © Google's mobility indices between February and July, 2020 for four selected countries split by subjective groups.

this:
"These calculations have not been done to predict future concentrations, only to put the changes experienced between March and July, 2020 in context."

12. Line 265: and biogenic VOCs emissions.

    Done.

13. Figure A3. What are the measurement methods and data reliability of VOC concentrations?

    The observations displayed in Figure A3 were gained from the Automatic Hydrocarbon Network. This network uses automatic gas chromatographs to report real-time speciated hydrocarbons. These data are reported to the European Commission and are considered high-quality. For more information, please visit `https://uk-air.defra.gov.uk/networks/network-info?view=hc`.

Other changes

- The data file which was provided as a temporary link has been migrated to a persistent data repository (https://doi.org/10.5281/zenodo.4464734). This repository is now referenced in text and in the data availability section.
- Three additional references have been added which report air quality changes due to COVID-19 lockdown measures.
- Table 1 had a formatting error which resulted in duplicate rows. This has been fixed.

COVID-19 lockdowns highlight a risk of increasing ozone pollution in European urban areas

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Abstract.

In March 2020, non-pharmaceutical interventions in the form of lockdowns were applied across Europe to urgently reduce the transmission of SARS-CoV-2, the virus which causes the COVID-19 disease. The ~~near-complete shutdown of the~~ aggressive curtailing of the the European economy had widespread impacts on atmospheric composition, particularly for nitrogen dioxide (NO_2) and ozone (O_3). To investigate these changes, we analyze data from 246 ambient air pollution monitoring sites in 102 urban areas and 34 countries in Europe between February and July, 2020. Counterfactual, business as usual air quality time series are created using machine learning models to account for natural weather variability. Across Europe, we estimate that NO_2 concentrations were 34 and 32 % lower than expected for traffic and urban-background locations while O_3 was 30 and 21 % higher (in the same environments) at the point of maximum restriction on mobility. ~~The European urban experienced in the~~ To put the 2020 changes in context, average NO_2 trends since 2010 were calculated, and the changes experienced across European urban areas in 2020 ~~lockdown~~ was equivalent to ~~that~~ 7.6 years of average NO_2 reduction (or concentrations which might be anticipated in 2028~~based on average trends since 2010. Despite~~). Despite NO_2 concentrations decreasing by approximately a third, total oxidant (O_x) changed little, suggesting that the reductions of NO_2 were substituted by increases in O_3. The lockdown period demonstrated that the expected future reductions in NO_2 in European urban areas are likely to lead to ~~a widespread increase in urban~~ widespread increases in urban O_3 pollution unless additional mitigation measures are introduced.

1 Introduction

On December 31, 2019, a cluster of unexplained pneumonia cases in Wuhan, Hubei, China was reported to the World Health Organization (WHO) (World Health Organization (WHO), 2020a; Wu et al., 2020). Subsequent research in January, 2020 identified the disease to be caused by a previously unknown betacoronavirus (SARS-CoV-2), and the disease was given the name coronavirus disease 2019 (COVID-19) (Zhou et al., 2020; World Health Organization (WHO), 2020c). Due to rapid human-to-human transmission and the introduction of the virus to countries outside China, cases of COVID-19 were soon detected in all continents of the world, with the exception of Antarctica, and on March 11, the WHO declared a COVID-19 pandemic (World Health Organization (WHO), 2020b).

Europe was named the epicentre of the pandemic on March 13, and most European countries undertook unprecedented non-pharmaceutical interventions to reduce the transmission rate of SARS-CoV-2 in early or mid-March (BBC, 2020; Dehning et al., 2020; Remuzzi and Remuzzi, 2020). The exact nature and duration of the measures varied by country, but collectively they are often referred to as "lockdowns" (Ruktanonchai et al., 2020). The lockdowns generally resulted in the closure of all shops, schools, universities, and restaurants with the exception of supermarkets, pharmacies, and other services deemed essential. Working from home whenever possible was encouraged and some countries also controlled, or restricted travel, exercise, and leisure activities. All these measures created a situation where European economic activity was reduced to a bare minimum within a matter of days ~~,~~ and mobility of the European population was severely altered. © Google's mobility data (© Google, 2020) based on movement trends very effectively demonstrates the change in mobility based on a baseline (Figure 1).

The rapid reduction of economic activity had many positive environmental impacts with the improvement of air quality being widely reported, especially via striking satellite observations of column NO_2 (Liu et al., 2020; Patel et al., 2020; Venter et al., 2020). Reductions of CO_2 emissions have also been reported globally due to heavily curtailed economic activities (Le Quéré et al., 2020; Forster et al., 2020). Many of the reports of improved air quality were preliminary, and further research was required to fully understand and quantify the improvements observed throughout Europe, particularly after accounting for meteorological factors ~~(Grange et al., 2020; Carslaw, 2020; Lee et al., 2020; Wang et al., 2020)~~(Grange et al., 2020; Carslaw, 2020; Lee et al., 2020; W
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The European lockdowns can be thought of and approached as an air quality 'experiment' where economic activity was ~~curtailed to near-minimum levels~~substantially curtailed where commercial, transportation, and recreation activities drastically declined. Questions can be asked from the data such as: what were the results, how do they compare to other planned interventions such as low emission or clean air zones, and whether the observations were inline with what would be expected? The rate and severity of the changes imposed on European populations due to the lockdowns is something that previously could only be investigated by atmospheric modeling. Therefore, the COVID-19 lockdowns have provided a unique 'real-world modeling scenario' which represents a plausible future with far fewer internal combustion engine vehicles in use across Europe.

Here, we report an analysis based on counterfactual business as usual scenarios using predictive machine learning models. This allows for robust comparisons of the observed concentrations of air pollutants with those which would have been expected without the lockdown measures. The primary objective of this study is to report the response of ~~and~~ NO_2 and O_3 concentrations throughout European urban areas caused by mobility restrictions due to COVID-19 lockdown measures. A secondary objective is to outline the implications for European air quality management which the dramatic changes in population mobility exposed.

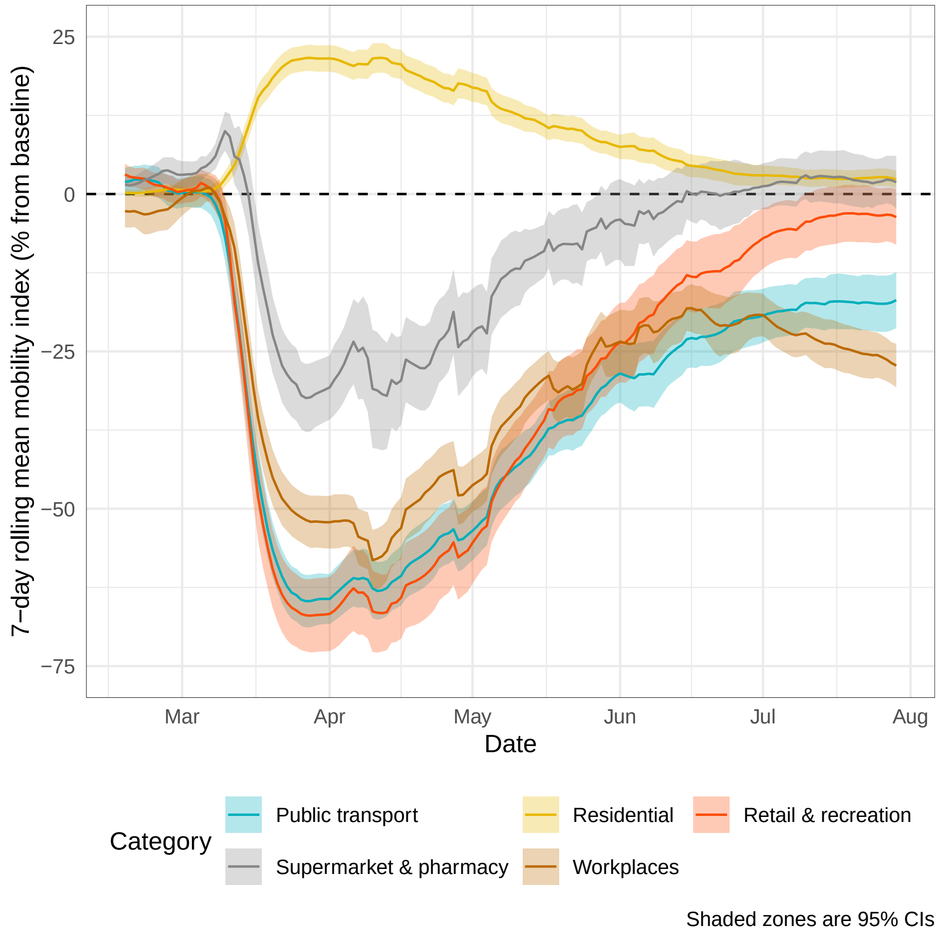

Figure 1. European mobility changes based on © Google's mobility indices between February and July, 2020 (© Google, 2020). The metrics display movement trends based on a baseline.

2 Materials and methods

2.1 Data

Up-to-date (UTD) hourly ~~and motioning~~ NO_2 and O_3 monitoring data were retrieved from the European Air Quality Portal (European Environment Agency, 2019) for the period between 2018 and 2020 for 102 urban areas in 33 European countries (Figure 2). For the 34th country, the United Kingdom, observations were directly retrieved from the countries' individual (England, Wales, and Scotland) and national networks (Automatic Urban and Rural Network; AURN) (Department for Environment Food & Rural Affairs, 2020).

The 102 urban areas were chosen because they are the capital, a "principal", or a particularly relevant city for the included European countries (Figure 2). In each urban area, at least one representative traffic site and at least one urban-background

Figure 2. The 102 European urban areas included in the data analysis.

site were chosen (if available) to represent the area. The mean distance among the different air quality monitoring sites within an urban area was 5.2 km. Notably, UTD data are not validated, are subject to change, and will only be finalised (at the time of writing) in ~~11~~ nine months time (the deadline is September, 2021). However, the time series were screened for undesirable features such as calibration issues, frequent missing data, or long periods of no reported data. Time series with such obvious issues were not included in the analysis. Unfortunately, oxides of nitrogen (NO_x = NO_2 + NO) data were not available because most countries which participate in the UTD process do not report NO_x (or NO) since it is not a regulated, ambient pollutant in Europe (Grange, 2019). Additionally, total oxidant (O_x = NO_2 + O_3) was calculated (in ppb) and included in the analysis as a third variable.

Hourly surface-based meteorological data were downloaded from the Integrated Surface Database (ISD). For the 102 urban areas, these sites were generally airports (NOAA, 2016; Grange, 2020). The matching logic between the air quality and meteorological sites was simple. The nearest ISD site to a particular air quality site was determined, the observations queried, and tested to ensure the data record was complete for the analysis period. If this criterion was met, the site match was positive and used for the analysis. A total of 246 air quality monitoring sites and 91 meteorological sites were included in the analysis. For details of the sites, see the ~~tables available online~~[3]table provided in an accompanying, persistent data repository (Grange, 2021).

In the current work, we focus on changes in the concentrations of ~~and~~ NO_2 and O_3 at urban-traffic and urban-background locations. ~~and~~ NO_2 and O_3 in such locations are strongly influenced by local road vehicle emissions and not, for example, transboundary contributions, which would be the case for particulate matter ($PM_{2.5}$ and PM_{10}). Furthermore, the concentrations of ~~and~~ NO_2 and O_3 in urban areas are strongly influenced by local meteorological effects. Generally, traffic sites are located in close proximity to roads, and pollutant concentrations are forced by local vehicular emissions. The urban-background classification is more varied, but can be thought as environments away from the immediate vicinity of roads and industrial facilities but are still located within an urban area.

2.2 Business as usual (BAU) modeling

A central issue when considering changes in atmospheric concentrations due to an intervention is whether the change is due to variations in meteorological conditions or emission source strength (Grange and Carslaw, 2019). This problem is widespread and affects time scales from hours to years. It is particularly important in 'before-after' studies where meteorological change, rather than changes in emission source strength, can easily dominate the variation in concentrations. This ambiguity can be somewhat reduced by averaging over several years to account for past inter-annual variability. However, this approach cannot account for the significant impact that meteorology may have on a specific observation period.

In the current context of the changes in activities brought about by COVID-19 lockdowns, the changes are over a duration of several months, and span a period from spring to summertime conditions. This period straddles important natural changes in meteorological conditions and atmospheric composition. For example, during February, 2020 the UK and much of western Europe experienced exceptionally high mean wind speeds due to storms Ciara, Dennis, and Jorge. Surface wind speed records in Southern England suggest February, 2020 had the highest mean wind speed of any month for over 40 years. This demonstrates that the state of the atmospheric dispersion across Europe at the time of COVID-19 lockdowns was different than experienced in previous years. Similarly, urban-background concentrations of O_3 in the northern hemisphere tend to increase from the beginning of the year and peak in April, which will also influence NO_2 (Monks, 2000). These, and other factors suggest that considerable care is needed for the quantification of an intervention such as the COVID-19 lockdowns on surface concentrations of primary and secondary pollutants.

To address the above issues, random forest models were trained to explain hourly mean ~~and~~ NO_2, O_3, and O_x concentrations using surface meteorological and time explanatory variables for each monitoring site (Breiman, 2001). The explanatory

[3]~~Temporary location:~~

variables used were: wind direction, wind speed, air temperature, relative humidity, atmospheric pressure (if available in the ISD database), a trend term in the form of Unix date, a seasonal term in the form of Julian day, weekday, and hour of day. The following random forest hyper-parameters were kept constant for all models: 300 trees, three variables to split at each node, and a minimal node size of five. The training period spanned just over two years and was between January 1, 2018 and February 14, 2020. The training-testing split percentage was 80 and 20 respectively. From February 14 to July 31, 2020, the models were used in predictive mode to predict pollutant concentrations based on the observed meteorological variables.

The ~~models' predictions can be thought of as business as usual (BAU) scenarios based on past behaviour of pollutant concentrations and the weather which was experienced after February~~philosophy of this approach involves using a machine learning model, trained on past data, to predict beyond the last observations it has seen. The model is trained on a substantially long period, two years in this work, to capture the variability of concentrations experienced in a variety of meteorological conditions. Beyond the training period (February, 14~~at each monitoring site. Thus~~, 2020), the model ~~represents~~ predicts concentrations based on meteorological variables which from the model's perspective are from the future. The time series which results is a *counterfactual*~~which observed~~. This counterfactual represents an estimate of concentrations during a business as usual (BAU) scenario. The BAU concentrations can be ~~compared with (~~readily compared with what was observed for example, ~~see~~ Figure 3 ~~)~~and the changes quantified, explained, and interpreted. This allows for a robust comparison with what was expected, with what was observed.

February 14 to March 1, 2020 was considered a validation period where the models' skill were checked for adequate performance. Summaries of the models' performance metrics based on the ~~random forest model objects and predictions during the validation period in the form of R^2~~ training and validation periods are shown in Figure A1. From the start date of the lockdowns (the earliest was March 9 in Italy), the application period began and gave estimates of BAU, *i.e.*, what concentrations would have been if the lockdown measures were not implemented. The modeling was conducted using the **rmweather** R package (Grange et al., 2018; Grange and Carslaw, 2019; Grange, 2018).

During the validation phase, a number of models showed bias in prediction, most notably, NO_2 was under-predicted at many locations. The under-prediction was on average ~~,~~ -3.7 $\mu g\, m^{-3}$ (95 % CI: [-4.2, -3.3]; mean percentage change: 15.9 %). This under-prediction was most likely caused by already-curtailed economic activity and reduced emissions throughout Europe at the very end of February and the beginning of March, *i.e.*, before the formal lockdowns were implemented. The beginning of 2020 was also mild in respect to ambient temperature and rather windy at most locations (discussed above) which may have resulted in some models under-predicting concentrations at this time of the year. For consistency and to create a reference point in time, the model predictions were corrected by calculating the model offset validation phase (February 14 to March 1) and subtracting this offset from the predictions. This ensured that the counterfactual predictions were calibrated at the start of the application phase and represented the changes in concentrations after March 1, 2020.

2.2.1 Change point analysis

To link ~~and~~ NO_2, O_3, and O_x concentration changes in March–April, 2020 to the lockdown restrictions placed on European populations, change point models were calculated. These change point models were conceptually simple – an intercept change

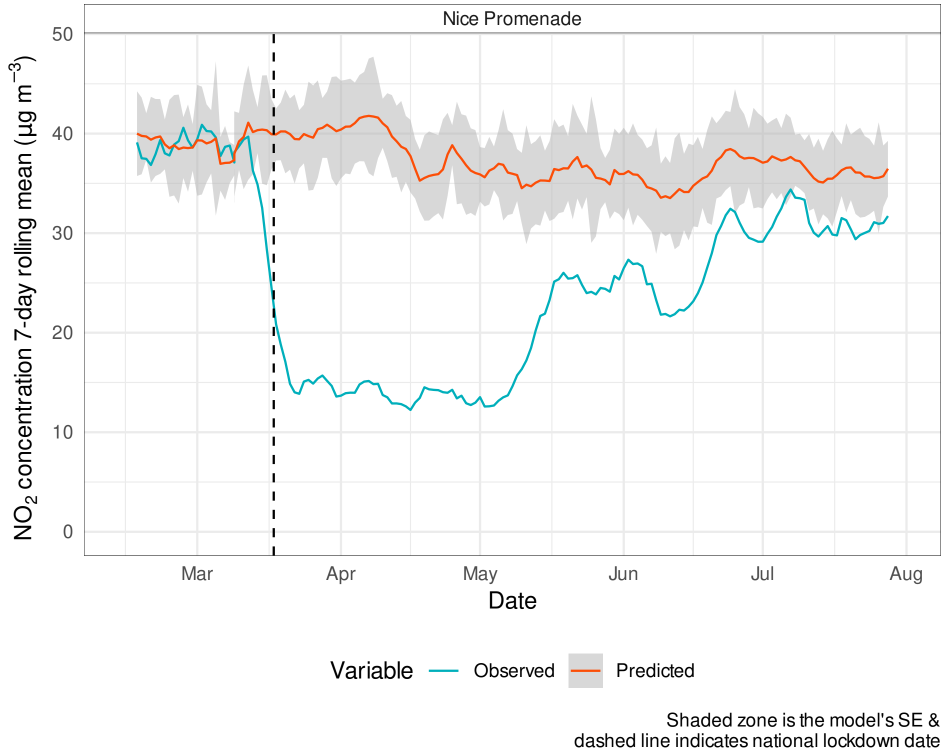

Figure 3. A NO_2 example where the observed concentrations clearly diverged from the business as usual (BAU) scenario for the Nice Promenade (France) traffic monitoring site between February and July, 2020.

was the expected *a priori* assumption. There were two motivations for these change point models. The first was to identify both the time, and magnitude of concentration response with an objective, data-driven approach rather than using a subjective and manual classifier. The second was to use such a technique to identify an atmospheric response after an intervention (an unplanned one in this case) which is a general goal of air quality data analysis.

The change point logic was implemented with the **mcp** R package with Bayesian inference (Lindeløv, 2020). To detect the change points, three Markov chains were run with 9000 iterations. The change point models tested the delta between the observed and counterfactual, however, the change-points were calibrated back to their pre-lockdown concentrations to conduct the (relative) percentage change calculations.

2.2.2 Presentation of results

When presenting the results of the analysis, most time series are displayed as seven-day rolling means. These rolling means act as a smoothing filter to make patterns clearer and remove the day-to-day variations generally seen in air quality monitoring data. Thirty-four countries were included in the analysis (Figure 2), but to avoid overwhelming plots and figures, a consistent set of six European countries (France, Germany, Italy, Spain, Switzerland, and the United Kingdom) were chosen to be displayed when discussing the counties' air quality patterns.

3 Results and discussion

3.1 Mean concentration changes

For all 34 European countries analysed, the observed concentrations of NO_2 were lower than those predicted by the counterfactual business as usual (BAU) scenarios between February 14 and July 31, 2020 (deltas (Δ) between the observed concentrations and predicted counterfactual shown in Figure 4). The reductions of NO_2 were greater in both an absolute and relative sense at the sites classified as either roadside or traffic environments compared to urban-background locations which can be explained by NO_2 being primarily a traffic-sourced pollutant (Grange et al., 2017). The impacts of vehicle-flow reductions during the lockdowns were more dramatic in the close proximity of roads when compared to more distant urban-background locations.

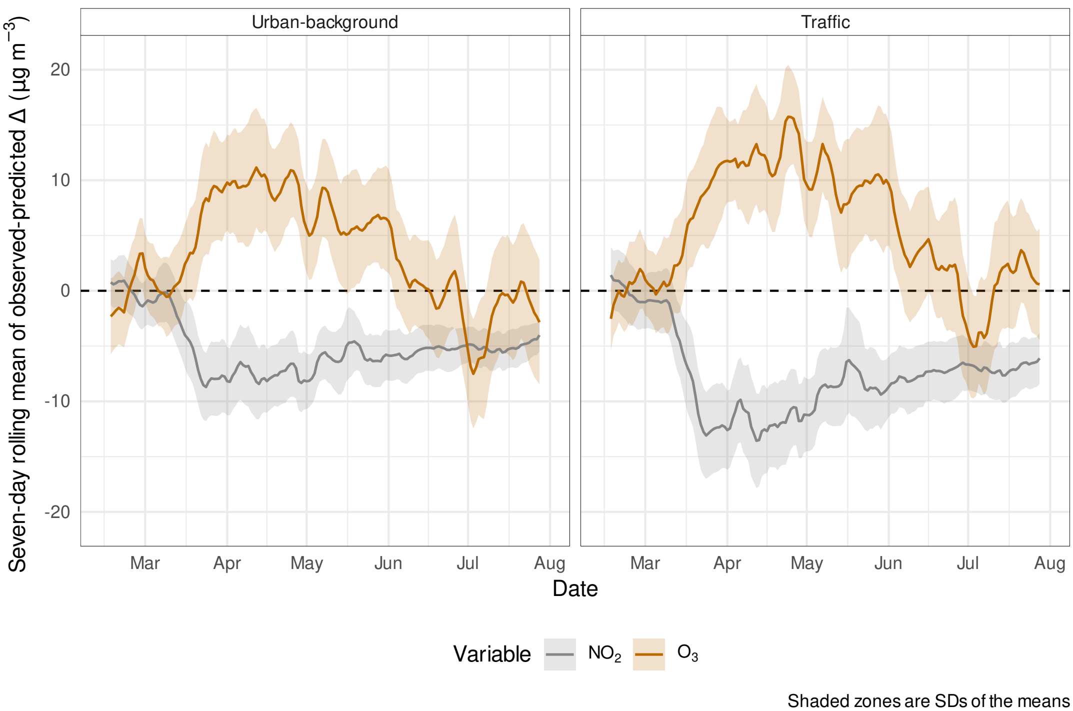

Figure 4. Seven-day rolling means of the observed-predicted concentrations deltas for NO_2 and O_3 for all European sites analysed between February 14 and July 31, 2020.

Mean O_3 concentrations increased at a similar magnitude to which NO_2 decreased throughout Europe between February and July, 2020 (Figure 4). Like ~~,~~ NO_2, O_3 at roadside locations showed a greater divergence from the BAU predictions than urban-background sites. The near-mirror image of ~~and~~ NO_2 and O_3 can be explained by the relationship between ~~and~~ NO_x and O_3.

The reduction of NO_x emissions and concentrations across Europe drove decreased O_3 destruction via the NO titration cycle during this period. In many countries, the 8-hour legal limit for O_3 of $120\,\mu g\,m^{-3}\;8\,h^{-1}$ was breached during this time period. Unlike NO_2 where concentrations remained below their BAU estimates until the end of the analysis period, O_3 concentrations returned to their expected values by the end of July, 2020.

3.2 Timing of changes

Figure 4 clearly indicates that concentrations in the first half of 2020 diverged from what was predicted by the counterfactual modelling. To objectively identify the date and magnitude of maximum divergence, change points were identified with a data-driven approach using Bayesian inference. The mean dates when NO_2 started to diverge at their greatest extent from the BAU scenarios along with national lockdown dates for six European countries are displayed in Figure 5. For the complete set of dates for all countries included in the analysis, see Table A1.

For NO_2, the change points were between seven days before and seven days after the countries' lockdown date (excluding the outlier of Denmark). For O_3, this range was greater, between -12 and 8 days. Italy was the first country in Figure 5 where change points were identified for NO_2 concentrations on March 13, 2020 and this was four days before Italy's nationwide lockdown date while Spain's NO_2 change point was the same as the country's lockdown date. Change points were often identified a day or two earlier than the lockdown date when the lockdown began on a Sunday or a Monday, for example, in Germany. For almost every site included in the analysis, the change points for NO_2 were ones of decreases while those for O_3 were increases (as seen in Figure 4).

Figure 5 shows that some countries had very consistent changes in concentrations for the sites which were analysed, for example Spain. Changes in other counties were less consistent which may indicate regional differences within countries. The UK showed two peaks in density for the NO_2 change points which were separated by a week. This feature represents a two-phase reduction in emissions because staggered lockdown measures were announced – the first was a set of recommendations for social distancing and not visiting restaurants and other social establishments (on March 16), while the second announcement (March 23) was one of a more strict lockdown.

Although the identified change point dates for NO_2 were broadly consistent with the various countries' lockdown dates, the change points for O_3 were not aligned as closely (Table A1). There was also no correlation between the magnitude of NO_2 reduction and the time required for an O_3 change point to be identified. This suggests that O_3's secondary generation processes did not immediately respond to reductions of ambient NO_x concentrations after lockdowns were imposed due to less NO titration. For this process to be identifiable, O_3 generation must occur, and this requires sunlight. Therefore, the lack of sunny conditions in some urban areas around the time of the NO_2 atmospheric response may have resulted in varying duration lags before changes in O_3 could be observed.

3.3 Concentration changes among different countries

At a European level, maximum divergence of ~~and~~ NO_2 and O_3 from the counterfactual predictions was reached in late-March, 2020 (Figure 4). However, there was some diversity among European country ~~and~~ NO_2 and O_3 divergence from their counter-

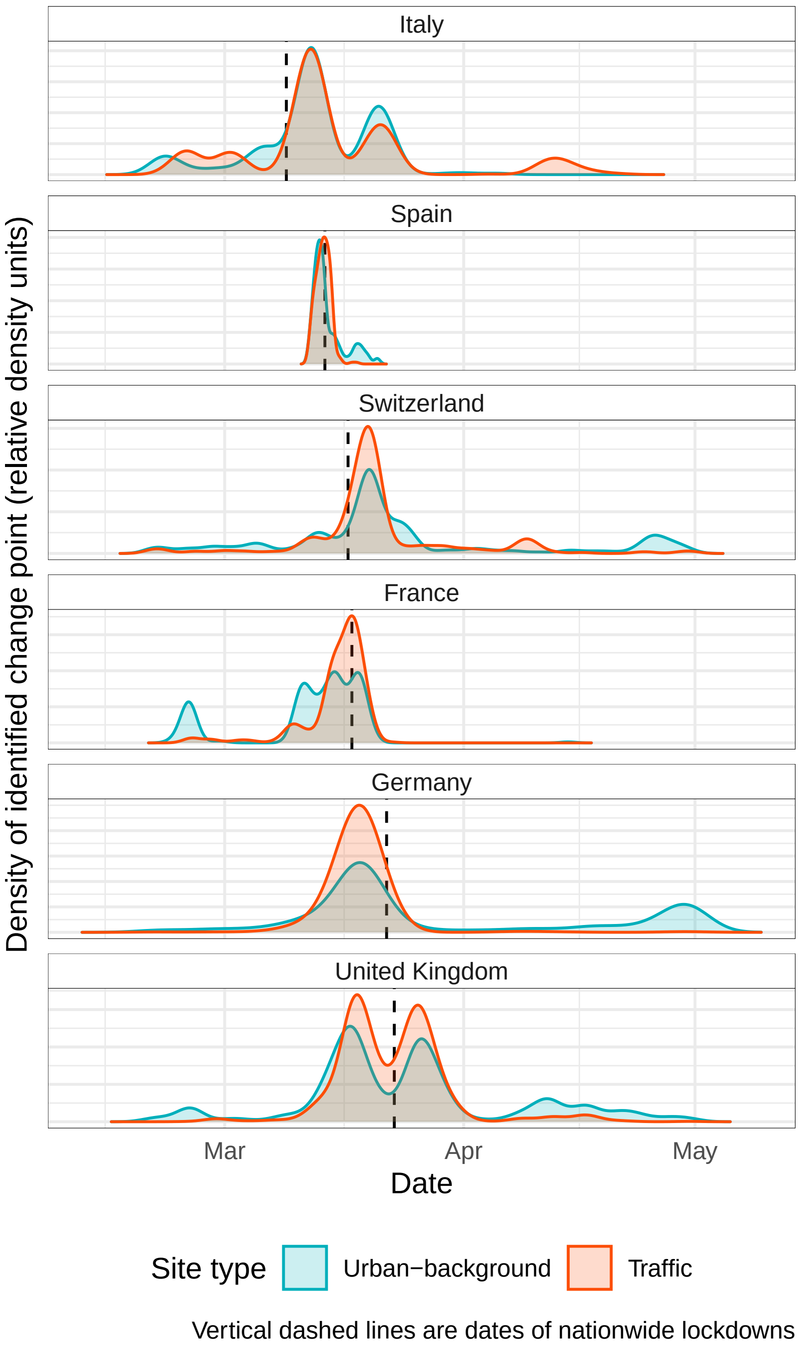

Figure 5. Estimated timing of changes to NO_2 concentrations for six European countries between March and May, 2020. The distribution shown for each country is the dimensionless probability distribution of the estimated change-point in concentration. The country panels are ordered by nationwide lockdown date.

factuals for the analysis periods (Figure 6). All countries analysed passed their maximum divergences for ~~and~~ NO_2 and O_3 in late-April, and the shape of the recovery is of a "swoosh" with a sharp plunge away from the counterfactual around the date of the lockdown implementations (Figure 6), but the rapid plunge was followed by a slower, and more gradual return to the BAU until the end of July. This pattern is very much reminiscent of the mobility changes shown in Figure 1.

Some countries experienced a smaller reduction in NO_2 than others. Germany and Switzerland for example, experienced lower NO_2 reductions when compared to France, Italy, and Spain. Some countries' greater reductions in ambient NO_2 concentrations could be explained by the level of "stringency" of the countries' lockdowns and resulting changes in mobility (Hale et al., 2020; © Google, 2020). For example, Germany and Switzerland's measures were very strong recommendations with few legally enforceable restrictions on recreational or leisure activities, while France, Italy, and Spain had more stringent requirements where movement and travel were restricted and enforced in a much stronger manner. It is very likely that these different levels (or enforcement) of restrictions had implications for emissions of atmospheric pollutants. However, meteorological conditions, perhaps similar synoptic scale patterns likely played a role in the differences observed among the countries too.

After late-April, concentrations moved towards their predicted counterfactual values and this continued to the end of the analysis period (Figure 6). Some European countries began to remove lockdown restrictions in the second half of April which increased traffic-sourced emissions, and this is consistent with the observations in Figure 4 and Figure 6. O_3 concentrations returned to approximately their BAU levels by the end of July, but NO_2 had yet to do so at the end of the analysis period, with the exception of Italy. This indicates that NO_x emissions (mostly traffic-sourced) had not yet reached their estimated BAU levels by the end of July across most of Europe after the country lockdowns were released.

3.4 Quantifying the changes in concentrations

The change point dates identified by Bayesian inference shown in Figure 5 and Table A1 were used to classify the time series as pre-lockdown, within lockdown, or post-lockdown periods. With this classification, concentrations were compared to calculate concentration deltas and percentage changes. At a European level, the mean NO_2 percentage changes for NO_2 at traffic and urban-background sites were -34 % (95 % CI [-36, -31]) and -32 % (95 % CI [-35, -29]) respectively (which equalled concentration reductions of -11 and -7 $\mu g\,m^{-3}$). The European annual NO_2 standard is 40 ~~y^{-1}~~ $\mu g\,m^{-3}\,y^{-1}$, and the mean reduction of 11 $\mu g\,m^{-3}$ is 27 % of the legal limit (European Commission, 2019). For O_3, the mean European percentage change for traffic and urban-background sites were estimated at 30 % (95 % CI [26, 35]) and 21 % (95 % CI [18, 24]), and the concentration changes were 12 and 9 $\mu g\,m^{-3}$ respectively. The concentration deltas and percentage changes attributed to the European lockdown measures are listed by country and site type in Table 1.

~~To put these concentration changes into context, and trend analysis between 2010 and 2019 for the 246 sites was conducted. Based on the sites which had a complete data record, the mean trends were -1.44 and -0.72 y^{-1} for at traffic and urban-background locations, while trends in the same environments were 0.2 and 0.49 y^{-1}. Therefore, at the roadside, the mean reduction of across Europe due to the COVID-19 lockdown measures was equivalent to that of 7.6 years of continued concentration reduction, or~~

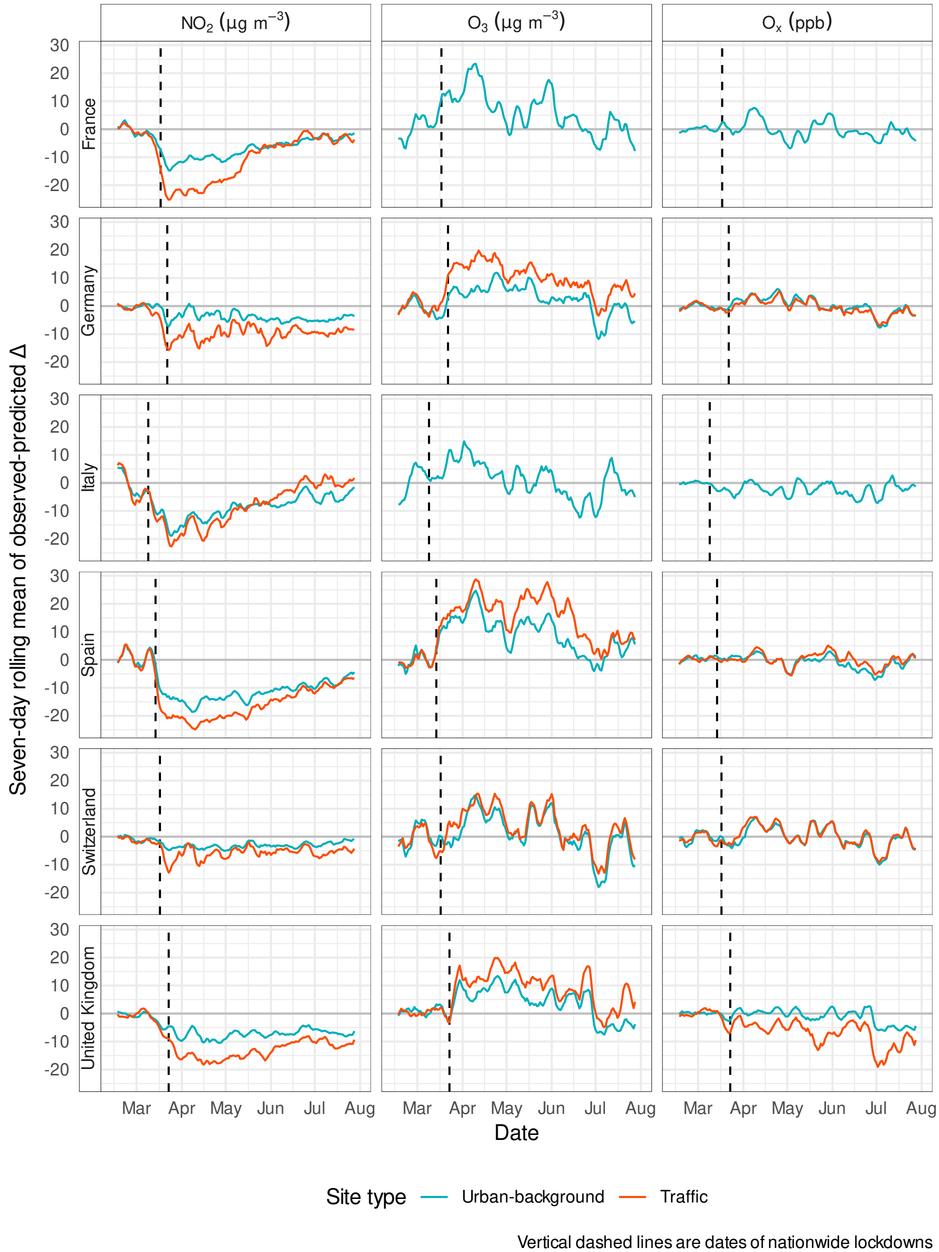

Figure 6. Seven-day rolling means of the observed-predicted concentrations deltas for NO_2, O_3, and O_x for six selected countries in Europe between February 14 and July 31, 2020.

~~equivalent to the anticipated European atmosphere in 2028 (). however, increased at an equivalent of 17 years of the rate of change determined by trend analysis in urban-background locations.~~

~~Mean European roadside trend with the reduction of concentrations attributed to the COVID-19 lockdowns put in context.~~

~~The changes at traffic sites will strongly reflect the influence of changes in traffic activity in close proximity to each site for , and . Close to roads, the origins of can be thought of as the combination of a background component, a component which is generated from the fast reaction between vehicular NO emissions and , and directly emitted (primary) . The primary contribution is known to have decreased in recent years from a peak around 2010. In London for example, the analysis of 35 traffic-influenced sites showed a reduction in the mean /vehicle emission ratio from around 25% in 2010 to about 15% in 2014, (~~Carslaw et al.~~, 2016) while at a European level, the /emission ratio peaked at 16 % (also in 2010) (~~Grange et al.~~, 2017). This decrease is believed to be driven by improvements in selective catalytic reduction control systems used on vehicles to reduce and also to the effect of ageing of diesel oxidation catalysts (~~Carslaw et al.~~, 2019).~~

~~The decrease in primary emissions over the past decade would have acted to reduce ambient concentrations close to roads. Indeed, if the traffic reductions experienced across Europe through country-wide lockdowns had occurred closer to 2010, the reductions in road vehicle emissions would have been much more important in affecting ambient concentrations than was experienced in early 2020.~~

To put these concentration changes into context, NO_2 and O_3 trend analysis between 2010 and 2019 for the 246 sites was conducted. Based on the sites which had a complete data record, the mean trends were -1.44 and -0.72 $\mu g\,m^{-3}\,y^{-1}$ for NO_2 at traffic and urban-background locations, while O_3 trends in the same environments were 0.2 and 0.49 $\mu g\,m^{-3}\,y^{-1}$. Therefore, at the roadside, the mean reduction of NO_2 across Europe due to the COVID-19 lockdown measures was equivalent to that of 7.6 years of continued concentration reduction, or equivalent to the anticipated European atmosphere in 2028 (Figure 7). O_3 however, increased at an equivalent of 17 years of the rate of change determined by trend analysis in urban-background locations. These calculations have not been done to predict future concentrations, only to put the changes experienced between March and July, 2020 in context.

The changes at traffic sites will strongly reflect the influence of changes in traffic activity in close proximity to each site for NO_x, NO_2 and O_3. Close to roads, the origins of NO_2 can be thought of as the combination of a background component, a component which is generated from the fast reaction between vehicular NO emissions and O_3, and directly emitted (primary) NO_2. The primary NO_2 contribution is known to have decreased in recent years from a peak around 2010. In London for example, the analysis of 35 traffic-influenced sites showed a reduction in the mean NO_2/NO_x vehicle emission ratio from around 25% in 2010 to about 15% in 2014, (Carslaw et al., 2016) while at a European level, the NO_2/NO_x emission ratio peaked at 16 % (also in 2010) (Grange et al., 2017). This decrease is believed to be driven by improvements in selective catalytic reduction control systems used on vehicles to reduce NO_x and also to the effect of ageing of diesel oxidation catalysts (Carslaw et al., 2019).

The decrease in primary NO_2 emissions over the past decade would have acted to reduce ambient NO_2 concentrations close to roads. Indeed, if the traffic reductions experienced across Europe through country-wide lockdowns had occurred closer to 2010, the reductions in road vehicle NO_2 emissions would have been much more important in affecting ambient concentrations than was experienced in early 2020.

The posterior draws (a type of model prediction) from the change point models show that in some countries, the ~~reduction~~ decrease of traffic volumes during the COVID-19 lockdowns reduced NO_2 concentrations to those which are experienced at

Table 1. Mean concentration deltas/differences and percentage changes of NO_2, O_3, and O_x for different countries and site types attributed to COVID-19 lockdown measures in March, 2020. Values which are missing indicates that there were not data and NC indicate no change point was identified.

		NO_2		O_3		O_x	
Country	Site type	Δ ($\mu g\,m^{-3}$)	% change	Δ ($\mu g\,m^{-3}$)	% change	Δ (ppb)	% change
Andorra	Traffic	–	–	–	–	–	–
Andorra	Urban-back.	-19.8	-59.7	16.1	43.0	-3.4	-9.8
Austria	Traffic	-7.6	-24.5	–	–	–	–
Austria	Urban-back.	-5.2	-23.1	11.3	19.5	4.3	11.2
Belgium	Traffic	-10.8	-45.3	5.0	10.5	-2.2	-6.5
Belgium	Urban-back.	-9.5	-38.4	8.9	19.2	2.4	6.5
Bosnia and Herzegovina	Traffic	–	–	–	–	–	–
Bosnia and Herzegovina	Urban-back.	-1.8	-11.9	1.4	15.0	-1.3	-3.4
Bulgaria	Traffic	-13.8	-29.5	14.0	29.6	0.9	2.2
Bulgaria	Urban-back.	-10.4	-34.2	13.9	33.6	3.0	8.4
Croatia	Traffic	-16.2	-42.3	–	–	–	–
Croatia	Urban-back.	-12.4	-43.9	21.5	34.1	4.4	9.6
Cyprus	Traffic	-15.3	-47.0	–	–	-2.8	-7.2
Cyprus	Urban-back.	-16.7	-59.7	6.1	10.9	-5.0	-11.8
Czechia	Traffic	NC	NC	–	–	–	–
Czechia	Urban-back.	NC	NC	9.0	18.3	4.9	13.8
Denmark	Traffic	-6.7	-28.0	15.7	31.7	3.9	9.8
Denmark	Urban-back.	-4.2	-49.0	7.6	12.3	3.1	8.4
Estonia	Traffic	-5.0	-35.2	0.7	1.3	-1.8	-5.2
Estonia	Urban-back.	-2.4	-29.2	6.4	10.7	-0.4	-1.2
Finland	Traffic	-9.4	-42.5	–	–	–	–
Finland	Urban-back.	-4.3	-34.1	–	–	–	–
France	Traffic	-20.3	-54.2	–	–	–	–
France	Urban-back.	-11.2	-44.1	13.9	35.0	-4.9	-12.1
Germany	Traffic	-10.5	-29.3	15.1	37.3	3.0	7.5
Germany	Urban-back.	-4.9	-21.6	8.8	16.6	3.5	9.1
Greece	Traffic	-12.3	-37.1	NC	NC	-1.1	-0.4
Greece	Urban-back.	-9.5	-43.9	NC	NC	-3.8	-8.5
Hungary	Traffic	NC	NC	–	–	–	–
Hungary	Urban-back.	NC	NC	5.0	15.7	-4.2	-11.4
Iceland	Traffic	-5.3	-33.7	–	–	–	–
Iceland	Urban-back.	-3.4	-23.5	–	–	–	–
Ireland	Traffic	–	–	NC	NC	–	–
Ireland	Urban-back.	-4.9	-33.6	NC	NC	-1.3	-3.5
Italy	Traffic	-17.3	-31.9	–	–	–	–
Italy	Urban-back.	-12.5	-32.7	3.8	14.1	-1.5	-2.2
Lithuania	Traffic	-7.0	-25.9	13.8	34.3	2.8	7.3
Lithuania	Urban-back.	-4.5	-21.0	–	–	–	–
Luxembourg	Traffic	-15.5	-53.2	–	–	–	–
Luxembourg	Urban-back.	-10.3	-47.0	9.6	17.0	-0.1	-0.3
Malta	Traffic	-13.2	-38.7	10.0	15.4	-4.1	-8.1
Malta	Urban-back.	–	–	–	–	–	–
Netherlands	Traffic	-6.2	-28.3	NC	NC	1.3	3.5
Netherlands	Urban-back.	-3.5	-21.2	NC	NC	4.1	11.2

Table 1. Continued.

		NO_2		O_3		O_x	
Country	Site type	Δ ($\mu g\,m^{-3}$)	% change	Δ ($\mu g\,m^{-3}$)	% change	Δ (ppb)	% change
North Macedonia	Traffic	-8.6	-33.2	NC	NC	-1.9	-6.8
North Macedonia	Urban-back.	–	–	NC	NC	–	–
Norway	Traffic	-7.7	-30.0	NC	NC	–	–
Norway	Urban-back.	-2.8	-17.1	NC	NC	0.9	2.2
Poland	Traffic	-11.7	-27.6	–	–	–	–
Poland	Urban-back.	-3.6	-12.7	7.1	15.1	2.1	5.5
Portugal	Traffic	-25.9	-53.8	20.2	46.8	-10.7	-24.6
Portugal	Urban-back.	-11.9	-40.5	13.8	26.8	4.7	12.1
Romania	Traffic	-5.8	-7.2	–	–	–	–
Romania	Urban-back.	-7.5	-26.3	13.0	39.9	-0.5	-0.5
Serbia	Traffic	–	–	–	–	–	–
Serbia	Urban-back.	-10.4	-56.4	15.6	44.9	-4.1	-12.6
Slovakia	Traffic	-6.8	-19.5	–	–	–	–
Slovakia	Urban-back.	–	–	–	–	–	–
Slovenia	Traffic	-9.6	-30.5	–	–	–	–
Slovenia	Urban-back.	-5.0	-18.9	20.9	55.7	8.2	26.1
Spain	Traffic	-22.8	-57.2	21.0	61.9	-1.5	-2.8
Spain	Urban-back.	-16.4	-55.7	15.9	37.5	-2.2	-5.4
Sweden	Traffic	-4.9	-17.0	–	–	–	–
Sweden	Urban-back.	-1.5	-12.5	6.5	12.2	0.6	2.0
Switzerland	Traffic	-5.5	-17.2	10.9	22.1	5.1	13.0
Switzerland	Urban-back.	-3.3	-10.1	11.7	21.7	5.2	14.4
United Kingdom	Traffic	-14.4	-50.8	14.4	45.8	-3.8	-8.3
United Kingdom	Urban-back.	-8.1	-36.8	8.0	16.4	0.0	0.1

urban-background locations (United Kingdom shown in Figure 8). The roadside increment ~~or enhancement of was essentially eliminated by reducing traffic to the levels which were experienced while in the lockdown state~~in NO_2 above urban background concentrations diminished considerably over lockdown due to large reductions in vehicle activity. However, as discussed above and shown in Figure 8, O_3 increased in response to the reductions of ~~and~~ NO_2 and O_x only altered very slightly. The same patterns in the United Kingdom were also experienced in other European countries such as France and Spain, but were not as clear for counties such as Switzerland and Germany (Table 1).

3.5 O_x – NO_2 and O_3 repartitioning

Figure 4 and Figure 6 demonstrate that NO_2 concentrations and emissions decreased throughout Europe due to the COVID-19 lockdown measures, especially at the roadside. However, the reduction of NO_2 was accompanied by an increase of O_3 at a similar magnitude and resulted in O_x showing little change despite the large reductions in traffic-sourced NO_2 (for example, Figure 8).

Mean European changes in O_x were variable between the two site environments. At traffic sites, O_x decreased by -1 ppb (-1.8 %; 95 % CI [-4, 0.7]) while in urban-background locations, O_x increased by 0.7 ppb (2.1 %; 95 % CI [-0.2, 4]). In the case of the traffic sites, the modest decrease of O_x can be partially explained by decreased emissions of primary NO_2 (Grange

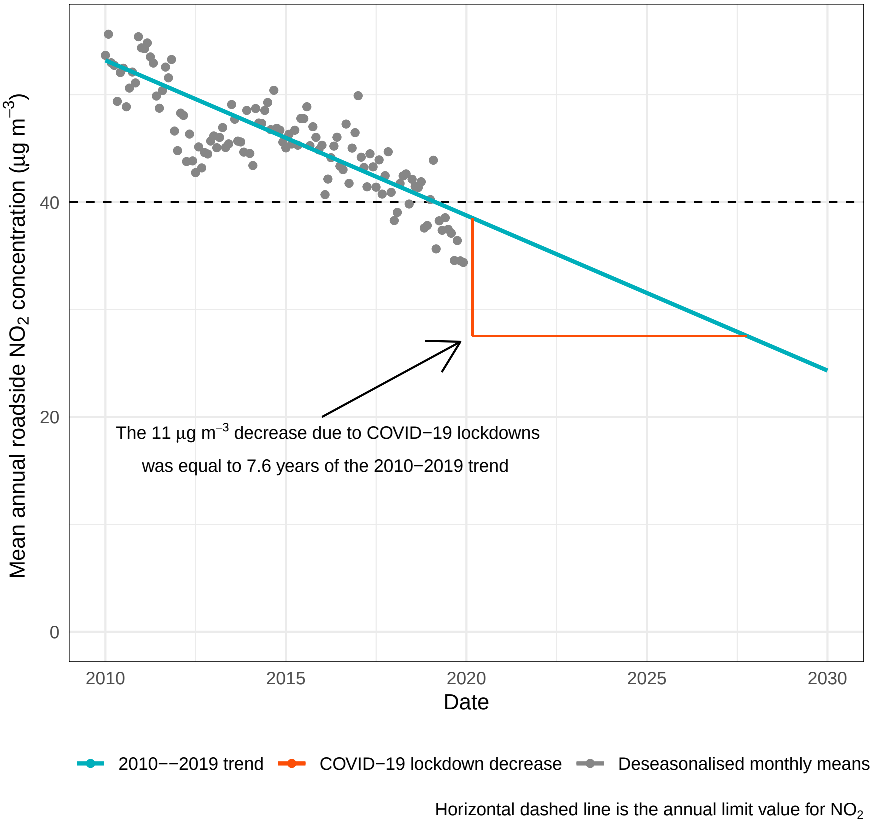

Figure 7. Mean European roadside NO_2 trend with the reduction of NO_2 concentrations attributed to the COVID-19 lockdowns put in context.

et al., 2017). However, in urban-background locations, O_x remained nearly constant. This is a very important observation for European air quality management. It suggests that the 34 % reduction of NO_2 concentrations was equalled by a similar absolute increase in O_3, which is clearly an undesirable outcome because of the deleterious effects of O_3 on population health, buildings, and vegetation.

The repartitioning of ~~to~~ NO_2 to O_3 is of importance from a public health perspective. As Williams et al. (2014) argue, there are good reasons from an atmospheric chemistry perspective to consider ~~and~~ NO_2 and O_3 together in epidemiological studies, rather than either of the two pollutants separately in single-pollutant models. Indeed, Williams et al. (2014) found that there were larger associations (on mortality) for mean 24 hour concentrations of O_x than for either ~~or~~ O_3 or NO_2 individually. On this basis, the current analysis suggest that the health impacts may have been small because O_x concentrations changed little in urban environments. The analysis conducted here was exclusively concerned with daily mean O_3 concentrations, and does not explore the subtleties associated with peak and/or increases in daily minima O_3 concentrations which are also important when considering the deleterious effects of O_3.

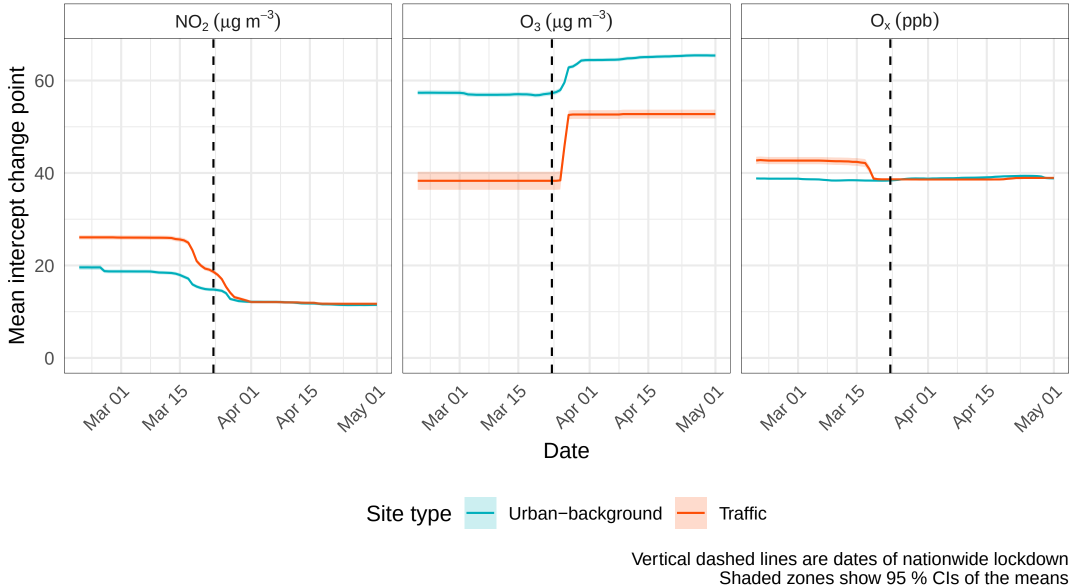

Figure 8. Posterior draws for NO_2, O_3, and O_x two-intercept change point models for the United Kingdom between March and May, 2020.

Efficacious management of O_3 has proven to be a challenge in Europe and in many other locations around the world (Sillman, 1999; Wang et al., 2017; Chang et al., 2017; Li et al., 2019). The struggle with O_3 control is partly due to the highly non-linear chemistry of O_3 production based on the precursors volatile organic compounds (VOCs) and NO_x. There are two regimes: NO_x-sensitive and VOC-sensitive – and the O_x analysis presented here strongly suggests that O_3 production is overwhelmingly VOC-sensitive across urban Europe. Therefore, if higher O_3 concentrations are to be avoided in the future where reductions in NO_x emissions of the scale seen in lockdown are likely, enhanced control of VOC emissions will be critical in the European urban environment. The prominence given to NO_2 as a pollutant following the dieselgate scandal of 2015 (Anenberg et al., 2017) has led to far more ambitious NO_2 emissions reductions policies in Europe than are currently in place for VOCs.

VOCs are only measured routinely in a few locations throughout Europe's urban areas, and represent a broad class of ~~pollutant~~ pollutants that are emitted from a wide range of sources. Whilst in the 1980s and 1990s VOC emissions were dominated by gasoline vehicle emissions (both tailpipe and evaporative), in more recent years their abundance has become increasingly influenced by non-transport sources such as natural gas leakage, biogenic emissions, and wider solvent use (Lewis et al., 2020).

Data from the London Eltham site, the only suburban VOC monitoring site in the UK, indicates that for many VOCs lockdown did not lead to significant changes in overall emissions or atmospheric concentrations (Figure A3). A conclusion from this albeit anecdotal evidence would be that further reductions in only traffic-related VOC emissions would not likely generate the desired air quality improvements in O_3 and that reducing emissions from other sectors would be essential.

Although out of scope for this current work, an obvious avenue for future research is to further explore how individual VOC concentrations responded during the lockdown periods in European urban areas in order to evaluate the proportion of VOCs that still come from traffic. This, combined with chemical modelling on a species by species basis to fully assess O_3 production chemistry, would help direct where future VOC reduction strategies should be focused. An analysis such as this would also strongly benefit from the access of NO_x (or NO as well as NO_2) data which, arguably, would be a better pollutant to analyse than NO_2 from an emissions perspective. We strongly encourage the institutions which are involved with reporting ambient air quality data to the European Environment Agency to include NO_x alongside the legally required NO_2 observations for the air quality community.

4 Conclusions

This work represents a classic air quality data analysis where atmospheric responses are linked to an intervention. In this case, the intervention was an unplanned, likely unique, and extreme event with very different characteristics when compared to typical interventions such as the introduction of new emission standards and low emission zones. Despite the extreme nature of the COVID-19 lockdowns and their results being much more impactful on urban atmospheric composition than other policies over a short time period, the analysis still demonstrates the difficulty of detecting “change upon change” for atmospheric pollutants – especially for locations where concentrations are close to background. However, this analysis presents a robust and portable framework for intervention analysis using a combination of machine learning-derived counterfactuals and change point analysis to identify the timing and magnitude of an atmospheric response.

Analysis of the effect of the European COVID-19 lockdowns on ~~, , and~~ NO_2, O_3, and O_x concentrations combining machine learning derived BAU modelling and Bayesian change point models indicate that NO_2 concentrations reduced by 34 % at roadside locations. However, the widespread reductions of NO_2 concentrations was accompanied by increases of O_3 at a similar magnitude (30 %), and thus, O_x altered only very slightly due to the lockdowns when considering Europe as a whole.

This insight has important implications for the implementation of future air quality management policies. The COVID-19 lockdown conditions give a glimpse of a realistic, and indeed likely, future environment where NO_x emissions continue to reduce at their current rate, primarily because of the increasing stringency of vehicular emission standards (Carslaw et al., 2016; Grange et al., 2017). The future reduction of NO_x concentrations will likely result in repartitioning of O_x and the increase of O_3 concentrations across most European urban areas. Although increases in European O_3 concentrations have been acknowledged, the further rise should be pre-empted by the European air quality management community through increased focus on VOC emission controls and the more holistic combined management of ~~,~~ NO_2, O_3, and VOCs. This will allow for continued improvements to air quality in a general sense, rather than focusing on reductions of individual pollutants.

Code and data availability. The data sources used in this work are described and some data sets are publicly accessible in a persistent data repository (Grange, 2021, https://doi.org/10.5281/zenodo.4464734). Additional data and information are available from the authors on reasonable request.

Author contributions. SKG and DCC conceived the research questions, conducted the analysis, and wrote the manuscript. JDL, WSD, and ACL contributed to the research design and writing of the manuscript. CH and LE reviewed and contributed to the manuscript writing.

Competing interests. The authors declare no competing interest.

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Table A1. Most commonly identified dates where observed and BAU modeled concentrations diverged in March, 2020. Dates which are missing indicates no change point was detected in March, 2020.

Country	Lockdown date	NO_2 date	O_3 date
Andorra	Fri., Mar. 13, 2020	Sat., Mar. 14, 2020	Thu., Mar. 19, 2020
Austria	Mon., Mar. 16, 2020	Thu., Mar. 19, 2020	Mon., Mar. 16, 2020
Belgium	Wed., Mar. 18, 2020	Sun., Mar. 15, 2020	Sat., Mar. 21, 2020
Bosnia and Herzegovina	Sat., Mar. 21, 2020	Thu., Mar. 19, 2020	Thu., Mar. 12, 2020
Bulgaria	Fri., Mar. 13, 2020	Wed., Mar. 11, 2020	Wed., Mar. 18, 2020
Croatia	Thu., Mar. 19, 2020	Fri., Mar. 20, 2020	Fri., Mar. 20, 2020
Cyprus	Sun., Mar. 15, 2020	Fri., Mar. 13, 2020	Thu., Mar. 19, 2020
Czechia	Mon., Mar. 16, 2020	–	Fri., Mar. 20, 2020
Denmark	Fri., Mar. 13, 2020	Fri., Mar. 27, 2020	Tue., Mar. 17, 2020
Estonia	Fri., Mar. 13, 2020	Mon., Mar. 16, 2020	Sat., Mar. 21, 2020
Finland	Mon., Mar. 16, 2020	Tue., Mar. 17, 2020	–
France	Tue., Mar. 17, 2020	Sat., Mar. 14, 2020	Wed., Mar. 11, 2020
Germany	Sun., Mar. 22, 2020	Sun., Mar. 22, 2020	Sat., Mar. 28, 2020
Greece	Mon., Mar. 16, 2020	Tue., Mar. 17, 2020	–
Hungary	Mon., Mar. 16, 2020	–	Sat., Mar. 14, 2020
Iceland	Mon., Mar. 16, 2020	Sat., Mar. 14, 2020	–
Ireland	Fri., Mar. 13, 2020	Thu., Mar. 19, 2020	–
Italy	Mon., Mar. 09, 2020	Fri., Mar. 13, 2020	Thu., Mar. 19, 2020
Lithuania	Mon., Mar. 16, 2020	Tue., Mar. 17, 2020	Wed., Mar. 11, 2020
Luxembourg	Mon., Mar. 16, 2020	Sat., Mar. 14, 2020	Fri., Mar. 20, 2020
Malta	Sun., Mar. 22, 2020	Sat., Mar. 14, 2020	Sun., Mar. 15, 2020
Netherlands	Mon., Mar. 16, 2020	Mon., Mar. 16, 2020	–
North Macedonia	Wed., Mar. 18, 2020	Fri., Mar. 13, 2020	–
Norway	Thu., Mar. 12, 2020	Tue., Mar. 17, 2020	–
Poland	Thu., Mar. 12, 2020	Tue., Mar. 17, 2020	Tue., Mar. 24, 2020
Portugal	Wed., Mar. 18, 2020	Sat., Mar. 14, 2020	Wed., Mar. 18, 2020
Romania	Mon., Mar. 16, 2020	Sat., Mar. 21, 2020	Tue., Mar. 17, 2020
Serbia	Sat., Mar. 21, 2020	Tue., Mar. 17, 2020	Mon., Mar. 16, 2020
Slovakia	Mon., Mar. 16, 2020	Sun., Mar. 22, 2020	–
Slovenia	Mon., Mar. 16, 2020	Thu., Mar. 12, 2020	Tue., Mar. 17, 2020
Spain	Sat., Mar. 14, 2020	Sat., Mar. 14, 2020	Sun., Mar. 15, 2020
Sweden	–	Wed., Mar. 18, 2020	Fri., Mar. 20, 2020
Switzerland	Tue., Mar. 17, 2020	Sun., Mar. 22, 2020	Thu., Mar. 26, 2020
United Kingdom	Mon., Mar. 23, 2020	Mon., Mar. 23, 2020	Thu., Mar. 26, 2020

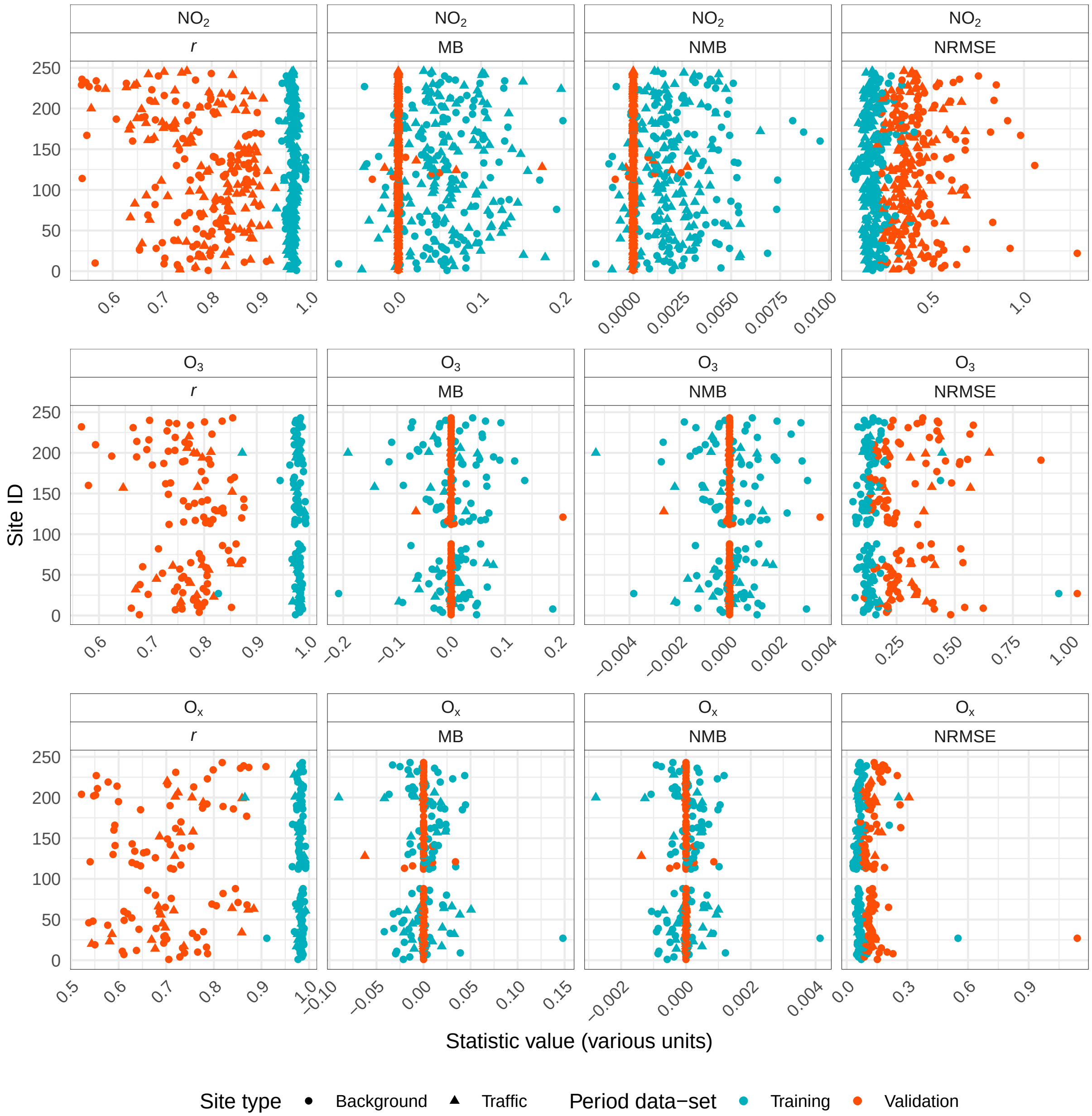

Figure A1. ~~Summaries of R^2 values from the random forest model objects~~ Model error summaries for all monitoring sites' (coded as integers) NO_2, O_3, and O_x models for ~~predictions during~~ two datasets – the ~~model~~ training and validation ~~period~~ sets. The error summaries are Pearson's correlation coefficient (~~February 14 to March 1~~ r), ~~2020~~mean bias (MB; in $\mu g\,m^{-3}$)~~for~~, normalised mean bias (NMB), and normalised root mean square error (NRMSE). The normalised were normalised by the ~~three predicted variables~~observed mean.

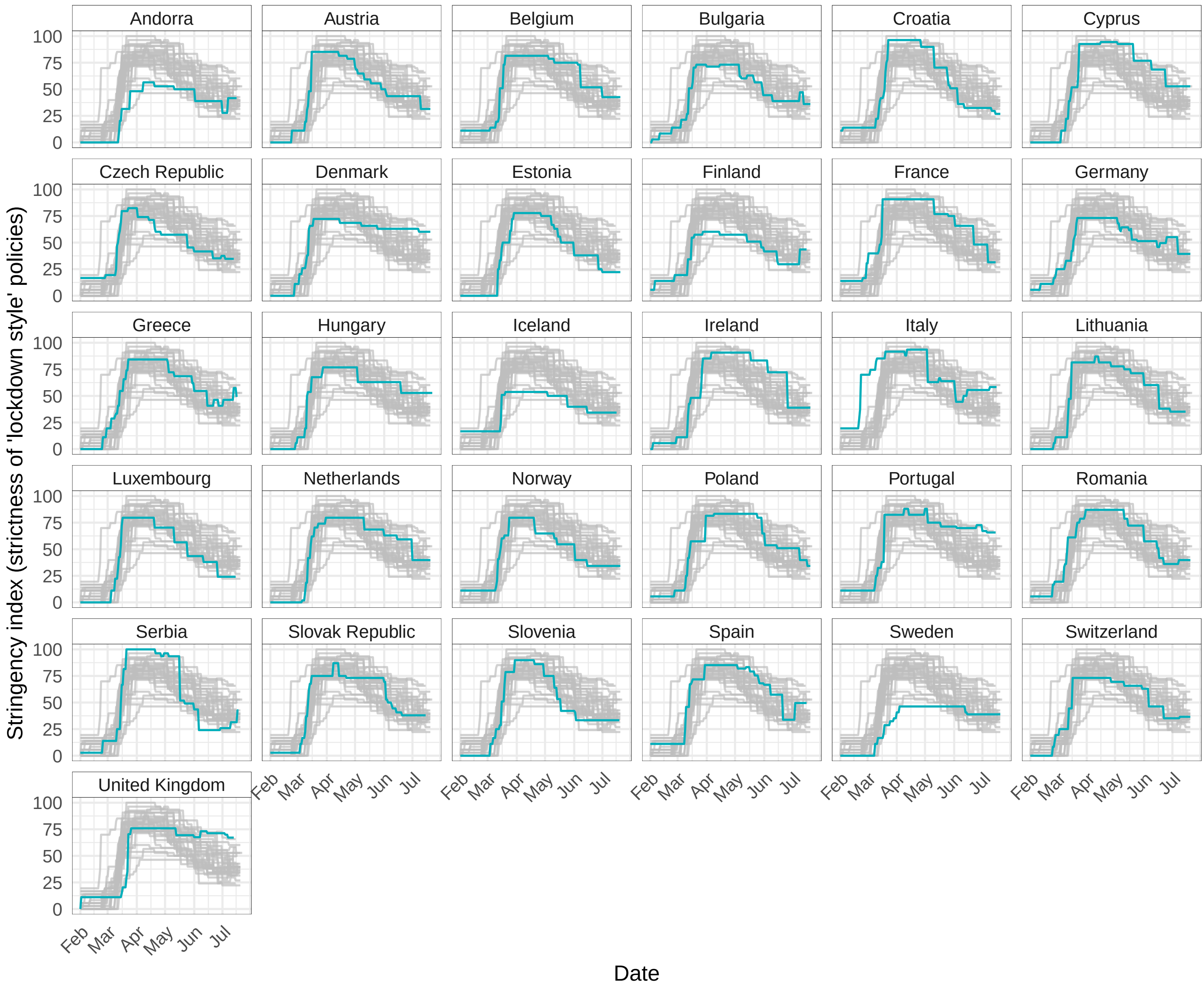

Figure A2. Oxford COVID-19 Government Response Tracker's (OxCGRT) stringency index of COVID-19 lockdown measures imposed by different countries' governments between February and July, 2020 (Hale et al., 2020).

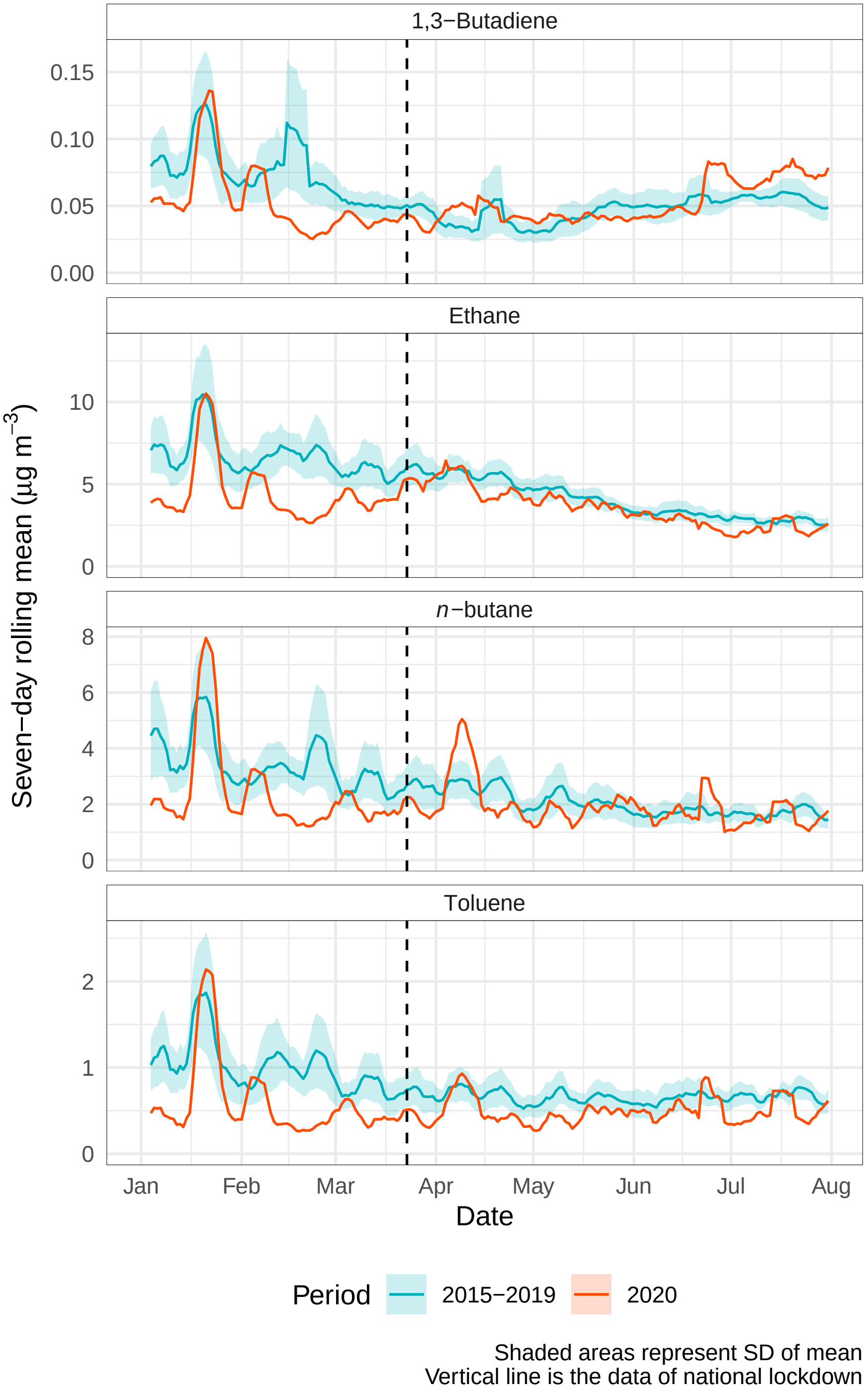

Figure A3. Volatile organic compounds (VOCs) time series at London Eltham, an urban-background site in United Kingdom.

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